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Data Article

Data sets of migration barriers for atomistic Kinetic Monte Carlo simulations of Cu self-diffusion via first nearest neighbour atomic jumps

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ABSTRACT

Atomistic rigid lattice Kinetic Monte Carlo (KMC) is an efficient method for simulating nano-objects and surfaces at timescales much longer than those accessible by molecular dynamics. A laborious and non-trivial part of constructing any KMC model is, however, to calculate all migration barriers that are needed to give the probabilities for any atom jump event to occur in the simulations. We have calculated three data sets of migration barriers for Cu self-diffusion with two different methods. The data sets were specifically calculated for rigid lattice KMC simulations of copper self-diffusion on arbitrarily rough surfaces, but can be used for KMC simulations of bulk diffusion as well.

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Specifications Table

Subject area	*Physics*
More specific subject area	*KMC simulations of surface diffusion*
Type of data	*Table*
How data was acquired	*Nudged elastic band calculations with semi-empirical potentials*
Data format	*raw*
Data source location	*Helsinki, Finland*
Data accessibility	*data is with this article*

Value of the data

- Cu Set 1 and Cu Set 2 tables of migration energy barriers can be used for atomistic rigid lattice Kinetic Monte Carlo simulations of Cu self-diffusion on arbitrarily rough surfaces via first nearest neighbour jumps.
- Bulk diffusion is also possible to simulate with these data sets in KMC.
- Cu Set 3 could be further statistically processed and analyzed.

1. Data

Cu Set 1 and *Cu Set 2* tables accompanying this article contain 5 columns labelled *a, b, c, d, Em*, where *a* and *b* are the numbers of first nearest neighbours (1nn) and second nearest neighbours (2nn), respectively, of the initial configuration of the jumping atom; *c* and *d* are the corresponding numbers for the final vacant lattice site (*c* includes the count of the jumping atom itself in the initial position); *Em* is the energy barrier in eV, which the jumping atom needs to overcome in order to make a transition from its initial configuration to the final vacant lattice site. We will henceforth call this the 4D parameterization scheme.

Cu Set 3 has the following format:

$$s0\ \ s1\ \ s2\ \ s3 \ldots s24\ \ s25\ \ Em$$

where *s0, s1, … s25* are the occupation states (1 = occupied, 0 = vacant) of the 26 closest (1nn and 2nn) sites around the jumping atom before and after the jump, *Em* is the corresponding barrier value in eV. We will henceforth call this the 26D parameterization scheme. All sets are described and analysed in detail in [1].

2. Computational methods

2.1. 4D and 26D parameterization schemes of rigid lattice atomistic Kinetic Monte Carlo models

Cu Set 1 and *Cu Set 2* were constructed within the 4D parameterization scheme of the KMC code Kimocs [2]. In the 4D description of the atomic jumps, only the numbers of 1nn and 2nn of the initial and final site of the transition are taken into account, but not the precise arrangement of these neighbours (see Fig. 1 for an example). Thus, a single value of the energy barrier is assigned to the whole set of various permutations corresponding to the same (*a*, *b*, *c*, *d*) 4D vector. Such an approach significantly reduces the set of necessary barriers from 2^{26} down to ~ 5000 for 1nn jumps in face-centred cubic (FCC) lattice structures.

The *Cu Set 3* table was constructed within the 26D parameterization scheme, i.e. taking into account not only the numbers of 1nn and 2nn atoms of the initial and final sites of the transition, (26 in total in FCC), but also the arrangement of these neighbours. In this scheme, if all barriers are to be calculated, then even in a mono-elemental metal 2^{26} barriers are needed. Fig. 2 shows the numbering

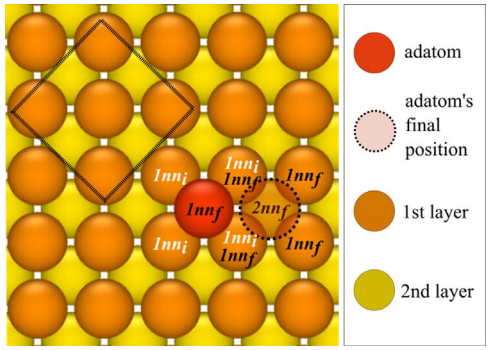

Fig. 1. Illustration of a (4,1,5,1) 1nn jump on the {100} FCC surface in 4D parameterization scheme. The adatom (red circle) jumps from the site with four 1nn atoms and one 2nn atom (the atom right below the jumping atom) to a site (dashed semi-transparent circle) with five 1nn (including the jumping atom itself) and one 2nn atom below it (marked with $\boldsymbol{2nn_f}$). To guide the eye, the FCC unit cell is shown with a square. Two surface layers are shown.

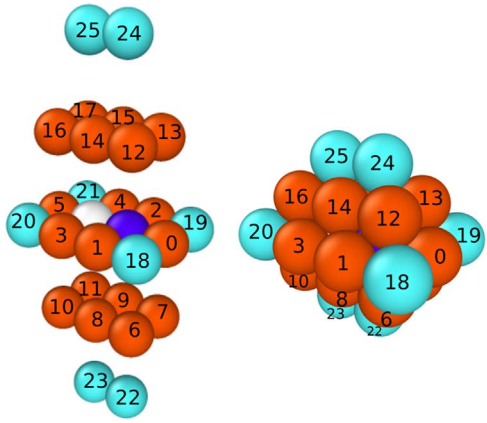

Fig. 2. The local atomic environment used in the 26D parameterization scheme: the octahedral atom cluster containing the migrating atom and its 1nn and 2nn sites (right); the indexing of sites from 0 to 25 within the cluster (left). The blue atom jumps to the vacant site shown with white color.

of the first and second nearest neighbours of the jumping atom and the final vacant lattice site that corresponds to the column order in *Cu Set 3*.

2.2. Migration barriers calculations with NEB in a rigid lattice

Migration barriers sets were calculated using the nudged elastic band (NEB) method [3,4] with the interatomic potential based on the Corrected Effective Medium Theory (CEM), developed by Stave et al. [5]. The initial and final configurations of every process were constructed in a rigid lattice, either on a surface or in a bulk (see [1] for details). During the initial relaxation stage, the initial and final configurations of processes were relaxed with the conjugate gradient method. The straight line from the relaxed initial position to the relaxed final one was chosen as the initial guess of the minimum energy path (MEP). The minimization of the interpolated path towards the MEP was handled by the NEB algorithm with Molecular Dynamics. The energy barrier is found from the relaxed MEP as a difference in the potential energies of the initial configuration and the configuration at the saddle point.

Surface relaxation effects in a rigid lattice in some cases may cause modifications of initial and/or final configurations during the initial relaxation stage and NEB calculations of energy barriers [1]. In these cases, the neighbourhood configurations of jumping atoms change either during the initial relaxations or NEB calculations. In other words, the intended event (a, b, c, d) in the 4D parameterization scheme becomes the event (a', b', c', d') and the barrier obtained in these calculations clearly cannot be used to describe the intended (a, b, c, d) event. The same problems may occur with the 26D parameterization scheme.

Some of such processes can be classified as *spontaneous*. A process is considered spontaneous if either its initial configuration relaxes towards its final configuration during the initial relaxation stage or there is no saddle point in the minimum energy path found during the NEB calculations. Such processes should happen immediately on real surfaces with no barrier. In the case of spontaneous events, the following heuristic formula was applied to calculate close to zero barriers:

$$E_m(a,b,c,d) = \epsilon a + \delta b + \epsilon c^{-1} + \delta d^{-1}, \quad (1)$$

where $\epsilon = 10^{-3}$ eV and $\delta = 10^{-4}$ eV. This formula is designed to prioritize the jumps of atoms with the fewest neighbouring atoms. It also assumes that it is more favourable for an atom to jump into a position with a higher number of neighbours. ϵ and δ are chosen so that the number of 1nn atoms contributes more into the value of migration barriers than the number of 2nn atoms.

Another type of processes that involves atoms in unstable configurations is more difficult to interpret than spontaneous events. In such processes, the jumping atom or its neighbouring atoms are either relaxed during the initial relaxation stage or dragged by NEB to some unrelated positions, which are different from the intended ones. These processes are rather unlikely to happen on real surfaces, but inevitable in the rigid lattice approximation. These processes present the biggest challenge in the calculation of the barriers. To address it, we used the tethering force approach [1]. During minimization, an additional spring tether is applied to all atoms in a simulation box. This spring force attracts atoms to the lattice sites. Depending on the strength of the spring tether, the atoms can deflect from their initial sites more loosely or more rigidly. In this manner, all the barriers, including the barriers for unstable configurations may, in principle, be calculated.

2.2.1. Cu Set 1

NEB calculations of *Cu Set 1* were done using the MD code PARCAS [6–8]. We used the approach described in [9] for the calculation of the additional NEB spring force between the images. A sequence of 40 images was used for every jump. The initial and final images were relaxed with the conjugate gradient method and then fixed during the NEB calculations. Some barriers were identified as spontaneous during NEB. All the processes with $a \leq 3$ (2486 events) were assigned with small barriers according to Eq. (1) to decrease the number of calculations.

This set includes the barriers for 4289 (a, b, c, d) events, most of which were calculated in the bulk. 190 events with $a > 3$ were classified as spontaneous during NEB calculations and assigned the barrier values according to Eq. (1). The permutations for each *Cu Set 1* event were chosen randomly.

No tethering or other restrictions applied during the NEB calculations were used, thus it was not possible to calculate all the $E_m(a,b,c,d)$ barriers. The attempt frequency was fitted to MD simulations of the flattening time obtained for Cu surface nanotips (see [2] for further details). The obtained value of the attempt frequency: $\nu = 7 \cdot 10^{13} s^{-1}$. Although the set is not complete, the sufficient amount of barriers was calculated to obtain a good agreement between the KMC and MD simulations in [2].

2.2.2. Cu Set 2 and Cu set 3

The Lammps MD package [10] was used for the NEB calculations with a climbing image [11] and an additional tethering force, with the tethering force constant set to 2.0 eV/$Å^2$, was applied. The NEB spring force was 1.0 eV/$Å^2$ and a total of 24 images were used for both *Cu Set 2* and *Cu Set 3*.

The energy barriers for 5103 (a, b, c, d) events were calculated in *Cu Set 2* both on a surface and in bulk (see [1] for details). For each jump, the permutation with the lowest sum of the energies of the initial and final states was chosen to represent the family of the (a, b, c, d) event. Only 283 barriers of jump events were assessed with the use of Eq. (1); the rest of the barriers were calculated with the tethering force approach. *Cu Set 2* is complete. The attempt frequency value was found by fitting of tip

flattening time to MD results as described in [2]. The set was used to reproduce the KMC simulations in [2].

Cu Set 3 includes 334,725 random 26D processes out of possible 1nn jumps in FCC structures. Eq. (1) was not used for spontaneous events; instead, they are marked with 0.0 barriers. The set is incomplete and has not been used for the KMC simulations.

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Transparency document. Supplementary material

Transparency data associated with this article can be found in the online version at https://doi.org/10.1016/j.dib.2018.01.066.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at http://dx.doi.org/10.1016/j.dib.2018.01.066.

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